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Comparison of Field, Laboratory, and Theoretical Estimates of Global Nitrogen Fixation by Lightning

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The various theoretical, laboratory, and field estimates reported in the literature for global nitrogen fixation by lightning have been calculated by using different lightning frequency values, channel lengths, and energy values. We have recalculated each of the individual estimates by using values of 7 km for channel length, two equivalent return strokes per lightning flash, and 100 flashes per second for global frequency, and a lightning energy input of 5×10^9 J per flash. The adjusted mean value is 72.2 ± 96.2 Tg N yr^{-1} for the theoretical category, 19.1 ± 10.0 Tg N yr^{-1} for the laboratory category, and 152 ± 59.9 Tg N yr^{-1} for the field category. Each of the three category adjusted mean NO_x global production rates is larger than previously reported contributions to the global N budget, making lightning potentially the single largest source.

1. INTRODUCTION

Nitrogen oxides include NO, NO_2, HNO_3, PAN, and other organic and inorganic nitrates have an important role in tropospheric chemistry. Some of the more important global sources of the primary nitrogen oxides, defined here as NO and NO_2 or NO_x, include fossil fuel combustion, biomass burning, oxidation of atmospheric ammonia, and atmospheric lightning. While nitrogen fixation by lightning has been estimated to be 17% of the global NO_x production [*Logan*, 1983], this value is highly uncertain, particularly in light of the recent work by *Franzblau and Popp* [1989].

The reported values of global NO_x production by lightning vary significantly, ranging from 1.2 to 220 Tg N yr^{-1} for estimates based on theoretical calculations, laboratory measurements, and field observations. The calculations by the individual researchers, however, used different values for lightning frequency, channel length, energy dissipated by each lightning flash, and other variables to arrive at the global NO_x production by lightning.

We investigated previous estimates of nitrogen fixation by lightning that were derived from theoretical calculations [*Tuck*, 1976; *Griffing*, 1977; *Chameides et al.*, 1977; *Chameides*, 1979; *Hill et al.*, 1980; *Dawson*, 1980; *Borucki and Chameides*, 1984; *Bhetanabhotla et al.*, 1985], laboratory experiments [*Chameides et al.*, 1977; *Levine et al.*, 1981; *Peyrous and Lapeyre*, 1982], and field observations [*Noxon*, 1976, 1978; *Kowalczyk and Bauer*, 1981; *Drapcho et al.*, 1983; *Franzblau and Popp*, 1989]. In this note we seek to intercompare the various reported estimates by using a standard set of variables to recalculate and thus adjust each of the cited individual estimates.

2. GLOBAL NO_x PRODUCTION BY LIGHTNING

The global NO_x production by lightning can be estimated with a simple equation,

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$$G = CPF \tag{1}$$

where G is the global NO_x production by lightning (g N yr^{-1}), P is the production rate (molecules per flash), F is the lightning frequency (flashes per second), and C is a conversion factor,

$$C = 7.35 \times 10^{-16} (\text{g N yr}^{-1})/(\text{molecules per second}) \tag{2}$$

Estimates of the contribution of global production of NO_x by lightning are based on theoretical calculations, studies of laboratory discharges, and field measurements of NO and NO_2 associated with thunderstorms. The discrepancy in the different estimates of global production is partially related to the uncertainty of channel length, the lightning frequency, and total energy dissipated by each lightning flash.

On the basis of recommendations of *Brook et al.* [1962], *Uman* [1969], and *Few* [1975], the channel length adopted in the aforementioned studies varied between 2 and 10 km. Lightning channels are typically longer than 5 km for tropical thunderstorms, where most lightning activity occurs [*Kowalczyk and Bauer*, 1981]. *Krehbiel et al.* [1983] reported that the typical channel length in Japan is 3 km versus 7 km in Florida. While recognizing that channel lengths can vary considerably, we here used 7 km to calculate the value of global NO_x production by lightning.

The global lightning frequency used in the aforementioned theoretical, laboratory, and field studies varied from 100 to 1600 strokes per second. Although the value of 100 flashes per second is commonly used, cloud-to-ground (CG) flashes are comprised of more than one return stroke. Some of the aforementioned studies used an average of four return strokes per flash, yielding 400 strokes per second. Because intracloud (IC) lightning is more frequent than CG flashes [*Uman*, 1986], others assumed that the average CG/IC ratio was 3:1 and arrived at 1600 strokes per second for global lightning frequency. However, CG strokes are argued to be about 10 times more energetic than IC discharges [*Kowalczyk and Bauer*, 1981], and the first return stroke of a CG flash

TABLE 1. Theoretical Estimates of NO_x Production by Lightning

Reference	p* 10^{16} Molecules J^{-1}	E, 10^5 J m^{-1}	L, 10^3 m	P, 10^{25} Molecules $Stroke^{-1}$	F, 10^2 Strokes s^{-1}	G, 10^{12} g N yr^{-1}	G_n, 10^{12} g N yr^{-1}
Tuck [1976]	1.8†	1.0	6.25	1.1	5	4.2†	6.6
Griffing [1977]	10	3.2	10	32†	1	23.5†	36
Chameides et al. [1977]	6 ± 1	1.0	5	3 ± 0.5†	16	35 ± 5	22 ± 4
Chameides [1979]	12 ± 4	1.0	5	6 ± 2†	16	70 ± 24	44 ± 15
Hill et al. [1980]	80†	0.1	5	4	1.5	4.4†	295
Dawson [1980]	6	0.15	10	0.9†	5	3.3	22
Borucki and Chameides [1984]	9 ± 2	0.8	5†	2.1 ± 0.5	1.75	2.6 ± 0.6	33 ± 7
Bhetanabhotla et al. [1985]	32†	0.1	5	1.6	1	1.2†	119
Mean						18.0	72.2
Standard deviation						24.3	96.2

Global production G is the product of the production rate P, the stroke frequency F, and the conversion factor $C = 7.35 \times 10^{-16}$ g N yr^{-1}/molecules per second. Adjusted global G_n production rate is the product of the molecular production P, the energy input (En = 3.6×10^5 J m^{-1}), the channel length (7000 m), the stroke frequency Fn = 200 strokes per second, and the conversion factor C.

*Production rate P is the product of the molecular production p, input energy E, and channel length L.

†Inferred values, not stated in the original paper.

is about four times as energetic as the subsequent strokes [*Guo and Krider*, 1982]. *Dawson* [1980] argued that all strokes associated with a particular flash occur along the same channel and suggested counting only the first, most energetic, return stroke. *Hill et al.* [1980] obtained an equivalent value of 1.5 ± 0.5 for the ratio of the energy of a typical three stroke flash to that of the initial stroke. *Sisterson and Liaw* [1990] presented arguments for using two equivalent return strokes per flash for NO_x production rate calculations that include IC flash production. Taking into consideration all the above factors, we used a value of two return strokes per flash and 100 flashes per second for worldwide lightning frequency.

The value of 10^5 J m^{-1} for energy input by a lightning discharge adopted by *Tuck* [1976], *Chameides et al.* [1977], *Griffing* [1977], *Chameides* [1979], and *Peyrous and Lapeyre* [1982] was based on the recommendation of *Uman* [1969]. *Hill* [1979] reevaluated previous electrical, optical, and acoustic estimates and recommended a typical value of 10^4 J m^{-1}. This recommendation was adopted by *Hill et al.* [1980], *Dawson* [1980], *Levine et al.* [1981], and *Bhetanabhotla et al.* [1985] in their calculations for the global NO_x production by lightning. *Borucki and Chameides* [1984] reviewed the literature to compare estimates of the total energy per flash derived from optical [*Connor*, 1967; *Krider et al.*, 1968; *Barasch*, 1970; *Mackerras*, 1973; *Turman*, 1978; *Guo and Krider*, 1982] and electrical [*Wilson*, 1920; *Malan*, 1963; *Connor*, 1967; *Uman*, 1969; *Mackerras*, 1973; *Berger*, 1977; *Hill*, 1979] measurements. On the basis of the assumption that the total flash energy was 1.75 times larger than that of the first return stroke, *Borucki and Chameides* [1984] obtained an average total flash energy of 4.0 (±2.7) × 10^8 J for both measurement categories combined, although individual researcher channel lengths had not been adjusted to a fixed value. We recalculated the total flash energy for *Malan* [1963], *Connor* [1967], *Krider et al.* [1968], *Uman* [1969], and *Mackerras* [1973] using our assumed channel length of 7 km and two return strokes per flash and obtained an average of 1.2 (±0.3) × 10^9 J. However, on the basis of the charge transferred per lightning flash obtained from return stroke peak current measurements and the potential difference calculated from a simplified charge distribution model of a thundercloud, *Uman* [1987] estimated the total energy dissipated in a lightning flash of 5 km channel length to be 10^9 to 10^{10} J. In this note we use a value of 5×10^9 J per flash to calculate global NO_x production by lightning. We have chosen a high-end value for our adjustments and will discuss this choice at the end of the discussion section.

2.1. *Theoretical Calculations*

The results from theoretical model calculations [*Tuck*, 1976; *Griffing*, 1977; *Chameides et al.*, 1977; *Chameides*, 1979; *Hill et al.*, 1980; *Dawson*, 1980; *Borucki and Chameides*, 1984; *Bhetanabhotla et al.*, 1985] listed in Table 1 vary from 1.2 to 70 Tg N yr^{-1}. These literature estimates are based on a channel lengths of 5 to 10 km, lightning energy inputs from 10^4 to 3.2×10^5 J m^{-1}, and lightning frequencies ranging from 100 to 1600 strokes per second. We recalculated the original estimates by using the values discussed in the previous section (channel length of 7 km, lightning energy input of 5×10^9 J per flash, and a global flash rate of 100 flashes per second with two equivalent return strokes per flash). These "adjusted" estimates are also summarized in Table 1. (The term "adjusted" is used here and throughout the remainder of the text to mean that an estimate was recalculated by using the set of variables discussed in the previous section). The adjusted global production of NO_x by lightning has a mean of 72.2 Tg N yr^{-1}, with a range of 6.6-295 Tg N yr^{-1}. The shock wave model used by *Tuck* [1976] and *Chameides* [1979] resulted in lower values of production than the hot-channel model used by *Hill et al.* [1980] and *Bhetanabhotla et al.* [1985].

2.2. *Laboratory Experiments*

Measurements of NO_x production in laboratory discharges have been made by *Chameides et al.* [1977], *Levine et al.* [1981], and *Peyrous and Lapeyre* [1982]. The estimated global NO_x production by lightning calculated from the various measurements of laboratory discharges vary from 1.8 to 47 Tg N yr^{-1}. These estimates are based on a channel

TABLE 2. Estimates of NO_x Production by Lightning From Laboratory Experiments

Reference	Laboratory Spark, Joules	p, 10^{16} Molecules J^{-1}	E, 10^5 J m^{-1}	L, 10^3 m	P, 10^{25} Molecules $Stroke^{-1}$	F, 10^2 Strokes s^{-1}	G, 10^{12} g N yr^{-1}	G_n, 10^{12} g N yr^{-1}
Chameides et al. [1977]	3.6×10^{-2}	6 ± 1	1.0	5	3	16	35 ± 5	22 ± 4
	1350	8 ± 4	1.0	5	4	16	47 ± 23	30 ± 15
Levine et al. [1981]	$5–17 \times 10^3$	5 ± 2	0.1	10	0.5*	5	1.8 ± 0.7	18.5 ± 7.4
Peyrous and Lapeyre [1982]		1.6*	1.0	5	0.8*	16	9.6*	5.9
Mean							23.4	19.1
Standard deviation							21.2	10.0

Abbreviations are as in Table 1.
*Inferred values, not stated in original paper.

length of 5 to 10 km, energy inputs from 10^4 to 10^5 J m^{-1}, and lightning frequencies ranging from 500 to 1600 strokes per second. Again, for the purpose of comparison we adjusted the results by using a channel length of 7 km, a lightning energy input of 5×10^9 J per flash, and a global flash rate of 100 flashes per second with two equivalent return strokes per flash. Table 2 summarizes the laboratory studies of NO_x production by lightning. The adjusted global production of NO_x has a mean of 19.1 Tg N yr^{-1} with a range of 5.9 to 30 Tg N yr^{-1}.

2.3. *Field Measurements*

Estimates of NO_x production by lightning based on field measurements were given by *Noxon* [1976, 1978] at the Langmuir Laboratory, New Mexico, and at the Fritz Peak Laboratory, Colorado; by *Drapcho et al.* [1983] at Argonne National Laboratory, Illinois; and by *Franzblau and Popp* [1989] also at the Langmuir Laboratory. *Noxon* [1976, 1978] and *Drapcho et al.* [1983] obtained an estimate of 1×10^{26} to 4×10^{26} NO_2 molecules per flash, while *Franzblau and Popp* [1989] estimated 3×10^{27} NO_2 molecules per flash. Although the latter study reported 100 Tg N yr^{-1}, actual calculations from the information provided in their article yielded a value of 220 Tg N yr^{-1}, which has been confirmed by E. Franzblau (private communication, 1989). *Kowalczyk and Bauer* [1981] computed global NO_x production by using satellite measurements of CG and IC lightning frequency in conjunction with the *Noxon*'s [1976, 1978] estimated production of NO_2 molecules per equivalent return stroke. Details of our adjustment of the *Kowalczyk and Bauer* [1981] estimate are discussed in the next section. The estimated global NO_x production from field observations ranged from 5.7 to 220 Tg N yr^{-1}.

We adjusted the field observations by using our channel length (7 km) and flash rate (100 flashes per second). We also included the measured ratio of NO_2 to NO_x of 0.25 for the *Noxon* [1976, 1978] and *Franzblau and Popp* [1989] data and a ratio of 1 for the *Drapcho et al.* [1983] data. Table 3 summarizes the field studies and our adjusted computations. The estimated adjusted global NO_x production from field measurements has a mean of 152 Tg N yr^{-1}, with a range of 74-220 Tg N yr^{-1}.

There are observations of elevated concentrations of NO in anvils of thunderstorms [e.g., *Ridley et al.*, 1987; *Chameides et al.*, 1987; *Dickerson et al.*, 1987]. The number of flashes (cloud to ground, intracloud, corona discharge, etc.) were not known in these cases. NO source (attributed to ensemble lightning effects) concentrations were made on the

TABLE 3. Estimates of NO_x Production by Lightning From Field Measurements

Reference	Location	Instrument	L, 10^3 m	NO_2/NO_x	P, 10^{25} Molecules $Flash^{-1}$	F, 10^2 Flash s^{-1}	G, 10^{12} g N yr^{-1}	G_n, 10^{12} g N yr^{-1}
Noxon [1976, 1978]	Fritz Peak Lab., Colorado	absorption spectrometer	2	1	15 ± 5	1	11 ± 4	154 ± 56
	Langmuir Lab., New Mexico	absorption spectrometer						
Drapcho et al. [1983]	Argonne Lab., Illinois	chemiluminescent analyzer	3	1	40	1	30	74
Kowalczyk and Bauer [1981]	global	satellite						
CG			7	1	10	0.5	3.8	77
IC			5	1	1	2.5	1.9	83
IC + CG							5.7	160
Franzblau and Popp [1989]	Langmuir Lab., New Mexico	absorption spectrometer and chemiluminescent analyzer	7	0.25	300	1	220	220
Mean							66.7	152
Standard deviation							102.8	59.9

Abbreviations are as in Table 1; in addition, CG is cloud-to-ground lighting, and IC is intracloud lightning.

TABLE 4. A Comparison of Theoretical, Laboratory, and Field Category Global NO_x Production Rates for Different Lightning Energy Inputs

	Energy Input = 4×10^8 J per flash				Energy Input = 5×10^9 J per flash			
Category	Maximum	Minimum	Mean	Standard Deviation	Maximum	Minimum	Mean	Standard Deviation
Theoretical	23.6	0.5	5.8	7.7	295	6.6	72.2	96.2
Laboratory	2.4	0.5	1.6	0.8	30	5.9	19.1	10.0
Field	220.	74.	152.	59.9	220	74.	152.	59.9

Global NO_x production rate in Tg N yr^{-1}.

basis of limited mixing and entrainment estimates. Only the *Chameides et al.* [1987] study provided an estimate of global N by lightning, with a value of about 7 Tg N yr^{-1}. Because of the way this estimate was derived, we are unable to calculate an NO production rate for a single flash in this or the other NO concentration references mentioned immediately above. Although we could not include these observations in our study, they do suggest much lower global NO_x production rates than indicated by our adjusted values in Table 3.

Both *Noxon* [1976, 1978] and *Franzblau and Popp* [1989] used an absorption spectrometer to measure the overburden of NO_2 via the scattered sunlight during thunderstorms. *Noxon* [1976, 1978] assumed that all of the NO produced by lightning was converted to NO_2 by reaction with ambient O_3. However, *Franzblau and Popp* [1989] used a chemiluminescent NO_x analyzer to measure the ratio of NO_2 to NO_x and obtained a value of about 0.25 near the flash. Because the *Noxon* [1976, 1978] measurements were made at the same location, we assumed that the ratio value of 0.25 was applicable to his data as well as to our adjusted value. *Drapcho et al.* [1983] measured both NO and NO_2 with a chemiluminescent NO_x analyzer at a location about 500 m from the flash and obtained a NO_2 to NO_x ratio of approximately 0.94. The very different NO_2 to NO_x values are not surprising because the locations have significantly different ozone concentrations (Chicago having the larger concentrations), which is a critical factor in the NO to NO_2 conversion rate.

3. DISCUSSION

The large variations among the adjusted estimates are due to several factors. Results were obtained with different techniques and in different environmental conditions. Adjustment removed some of these differences, but not all.

NO_x production rates derived from theoretical models exhibit the largest relative variance of the three categories. The large variation appears to be, in part, a result of the modeling of the physics of the thermal energy dissipation. The differences between these two models, which is beyond the scope of our paper, cannot be resolved by our study. The mean adjusted theoretical NO_x production rate is 72.7 Tg N yr^{-1}, with a standard deviation of ± 96.2 Tg N yr^{-1}.

Laboratory studies use low-current, low-voltage, and very short gaps between electrodes, as compared to actual atmospheric lightning. The amount of energy for a laboratory spark used varied from 3.6×10^{-2} to 1.7×10^4 J. The measured production rates were scaled up to atmospheric lightning energy input values in a linear fashion. Experimental uncertainties and the assumptions of linear scaling of processes from the laboratory to the ambient atmosphere all have considerable uncertainty, beyond the scope of this paper. Our study indicates that laboratory estimates are nearly all below the theoretical and field-measured production rates. The mean adjusted laboratory NO_x production rate is 19 Tg N yr^{-1}, with a standard deviation of ± 10.0 Tg N yr^{-1}.

Field measurements of NO_x avoid many uncertainties associated with lightning, such as breakdown currents, energy per flash, and linear functions that scale up laboratory spark values to atmospheric lightning values. However, there are logistical difficulties in obtaining lightning measurements. The average adjusted field NO_x production rate is 152 Tg N yr^{-1}, with a standard deviation of ± 59.9 Tg N yr^{-1}. Comparison of the various field-measured NO_x production rates after adjustment shows the largest values (74 to 220 Tg N yr^{-1}) but the least within-category variation.

The *Kowalczyk and Bauer* [1981] adjusted global NO_x production estimate was based on the *Noxon* [1976, 1978] molecular production rate and flash rates derived from satellite observations. This estimate was fundamentally different from the other estimates because of its independent use of CG and IC flash frequencies and the associated uncertainties. We used the *Noxon* [1976, 1978] NO_x molecular production rate (1.5×10^{26} molecules per flash), multiplied by the ratio of our normalized channel length to the *Noxon* [1976, 1978] channel length (7/2), and multiplied by the NO_x/NO_2 ratio (1/0.25) to obtain a value of 2.1×10^{27} NO_x molecules per flash for CG lightning. We derived the molecular rate for IC lightning from the CG lightning rate by dividing by the ratio of our normalized channel length to the IC lightning channel length (7/5), multiplying by the ratio of the energy of IC to CG flashes (1/3), and multiplying by 0.9 to correct for altitude. This resulted in an IC lightning NO_x molecular production rate of 4.5×10^{26} molecules per flash. Our adjusted global production is the conversion facto C (2) times the sum of the IC molecular rate, times the IC observed flash frequency (250 flashes per second) with the CG lightning molecular production rate, times the observed CG flash frequency (50 flashes per second) and is equal to 160 Tg N yr^{-1}.

Clearly, we chose an energy input value that was at the high end of such estimates. Choosing a more conservative value of 4×10^8 J per flash [e.g., *Borucki and Chameides*, 1984], results in a much lower theoretical and laboratory category averages, as shown in Table 4. Because lightning energy input is not used in the calculation of field category adjusted values, there is no effect to the category average. However, as shown in Table 4, the standard deviations of the theoretical and laboratory categories do not overlap the field category's. Using the larger energy input value of 5×10^9 J per flash results in an overlap of the laboratory and field categories with the theoretical category. Intrinsically, because all three categories are observing the same phenomenon, they should have reasonable agreement. We have not made any judgement about which category is "best," just that results should be similar. Our choice of the higher-end energy input value allowed all three categories to have essentially overlapping standard deviations; i.e., not statistically different.

In principal, all three categories are investigating the same phenomenon. However, the actual set of physical conditions and assumptions for estimate of each category is different. Hence a simple average of the mean of each category is not valid. On the basis of the work presented in Tables 1–3 we conclude that normalized NO production rate by lightning ranges between 9.1 Tg N yr^{-1} (the lowest category mean value minus its standard deviation) and 211.9 Tg N yr^{-1} (the highest category mean value plus its standard deviation). However, for a best estimate of a single value for comparison to *Logan*'s [1983] single global NO_x production rate value (8 Tg N yr^{-1}) frequently quoted in the literature, we have used a simple average of the three category means and calculated the standard deviation as the square root of the some of the squares of the variance (standard deviation squared) for each category, to obtain a value of 81 ± 65.7 Tg N yr^{-1}. Overall, our work suggests that a single value estimate of 8 Tg N yr^{-1} for a global NO_x production rate may be low by a factor of 10.

4. CONCLUSIONS

The various theoretical, laboratory, and field observations previously reported in the literature for global nitrogen fixation by lightning have all been calculated by using different lightning frequency values, channel lengths, and energy values. As a result, the estimates are difficult to compare. We have recalculated each estimate by using the assumptions that the average length of a flash is 7 km, one flash consists of two equivalent return strokes, lightning worldwide occurs at 100 flashes per second (200 return strokes per second), and lightning energy input if 5×10^9 J per flash. The adjusted mean value is 72.2 ± 96.2 Tg N yr^{-1} for the theoretical category, 19.1 ± 10.0 Tg N yr^{-1} for the laboratory category, and 152 ± 59.9 Tg N yr^{-1} for the field category. Our results indicate that each of the three category adjusted mean NO_x global production rates is considerably larger than previous estimates of about 8 Tg N yr^{-1} by, for example, *Logan* [1983]. Although there is considerable uncertainty associated with theoretical, field, and laboratory estimates, at the very least our work suggests that lightning is potentially the single largest contributor to the global N budget.

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